

**PATENT APPLICATION****IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of  
Masaki FUKUMORI et al.

Docket No: Q94896

Appln. No.: 10/579,216

Group Art Unit: 1713

Confirmation No.: 1179

Examiner: Karuna P REDDY

Filed: May 12, 2006

For: AQUEOUS LIQUID DISPERSION OF WATER AND OIL REPELLENT  
AGENT

**DECLARATION UNDER 37 C.F.R. § 1.132**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Takashi Enomoto, hereby declare and state:

THAT I am a citizen of Japan;

THAT I have received the master degree (department of chemical for materials)  
in 1986 from Mie University;

THAT I have been employed by DAIKIN INDUSTRIES LTD. since April 1,  
1986, where I hold a position as researcher, with responsibility for research works on  
the development of the synthesis of fluorine-containing compound and the development  
of water and oil-repellent; I was engaged to work for developing water and oil-repellent  
from 1986 to 1996; I worked in production department of fluorine-containing products  
between 1996 and 1999; again, I am engaged to work for developing water and oil-  
repellent from 1999 to now; and especially, I worked in USA (DAIKIN AMERICA,  
INC.) between 2000 and 2006; and

THAT I am familiar with the Office Action dated July 31, 2008.

I report below on certain experimentation that was conduct by me or under my  
direct supervision.

## EXPERIMENTATION

The following Examples 1 to 10 and Comparative Examples 1 to 3 correspond to respectively Examples 1 to 10 and Comparative Examples 1 to 3 described in the present Description. Comparative Examples 4 and 5 correspond to respectively Examples 11 and 12 described in the present Description. This Declaration newly shows (i) Comparative Examples 6 and 7 and (ii) evaluation results of mechanical stability and chemical stability for Examples and Comparative Examples.

Evaluations are conducted in the following Examples and Comparative Examples as follows:

### Wash Durability of Water- and Oil-repellency

An aqueous dispersion is diluted with tap water to the solid content of 1.0% by weight, to prepare a treatment liquid. A cotton cloth is immersed in the treatment liquid, squeezed with a mangle to give a wet pick up of 60%, dried at 100°C for 2 minutes, and heat-treated at 160°C for 1 minute. Washing of AATCC-135 method is repeatedly conducted three times and then the water- and oil-repellency of the treated cloth (HL-3) is evaluated.

Water repellency: AATCC-22 method

Oil repellency: AATCC-118 method

### Storage Stability

The average particle diameter (scattering intensity) of an aqueous dispersion (solid content: 30% by weight) is measured at the initial and after the storage at 50°C

for one month by FPAR-1000 manufactured by Otsuka Electronics Co., Ltd. and the storage stability is evaluated in the following criteria:

Good: Change rate of smaller than 10%

Fair: Change rate of 10% to 20%

Poor: Change rate of larger than 20%

#### Mechanical Stability

An aqueous dispersion is diluted with tap water to a solid concentration of 0.2 % by weight and stirred with a homomixer at 3,000 rpm for 10 minutes. The generated scum is filtered off with a black cotton cloth.

Good: No scum generated

Fair: Slight scum generated

Poor: Much scum generated

#### Chemical Stability

An aqueous dispersion is diluted with tap water to a solid concentration of 0.6 % by weight. 0.03 % by weight of a fixing agent for nylon is added and the mixture is intimately mixed. The generation of agglomerated material is observed.

Good: No agglomerated material generated

Fair: Slight agglomerated material generated

Poor: Much agglomerated material generated

#### Yellowing of Cloth

An aqueous dispersion is diluted with tap water to the solid content of 1.0% by weight, to prepare a treatment liquid. A cotton cloth is immersed in the treatment liquid,

squeezed with a mangle to give a wet pick up of 60%, dried at 100°C for 2 minutes, and heat-treated at 160°C for 1 minute. A difference of the b value between the treated cloth and the untreated cloth is measured by CR-300 Color Difference Meter manufactured by Minolta Co., Ltd. and evaluated in the following criteria:

Good:  $\Delta b$  of smaller than 0.1

Fair:  $\Delta b$  of 0.1 to 0.5

Poor:  $\Delta b$  of larger than 0.5

#### Example 1

Into a 1 L autoclave,  $C_nF_{2n+1}CH_2CH_2OCOCH=CH_2$  (a mixture of compounds wherein n is 6, 8, 10, 12 and 14 (average of n: 8)) (FA) (a fluorine-containing monomer) (150 g), stearyl acrylate (75 g), N-methylolacrylamide (3 g), pure water (300 g), tripropylene glycol (80 g), polyoxyethylene lauryl ether (25 g) and an epoxidized soybean oil (10 g) were charged and emulsified by ultrasonic wave at 40°C for 30 minutes with stirring. After the emulsification, n-dodecyl mercaptan (1 g) was added and then vinyl chloride (a chlorine-containing polymerizable compound) (40 g) was injected. Further, azobisisobutyramidine dihydrochloride (0.8 g) was added and the reaction was conducted at 60°C for 5 hours to give an aqueous dispersion containing a polymer. The composition of the polymer was that the reaction conversion of vinyl chloride was about 80% and the reaction conversions of the other monomers were about 100%. Sodium hydrogen carbonate (0.7 g) was added to this aqueous dispersion to give an aqueous water- and oil-repellent dispersion having the adjusted pH of 7.

For the aqueous water- and oil-repellent dispersion, evaluated were washing durability at the initial and after storage at 40°C for one month, and the yellowing of cloth at the initial.

The results are shown in Table 1.

#### Examples 2 to 5

The same procedure as in Example 1 was repeated except that the same amounts of the compounds shown in Table 1 were used as the chlorine-containing polymerizable compound and the epoxy compound. In Example 4, the same amount (105 g) of stearyl alpha-chloroacrylate was used instead of vinyl chloride and stearyl acrylate.

The addition amounts of sodium hydrogen carbonate for change of an aimed pH value were as follows:

Example 2: 0.7 g

Example 3: 0.5 g

Example 4: 0.4 g

Example 5: 1.5 g

The results are shown in Table 1.

#### Examples 6 and 7

The same procedure as in Example 1 was repeated except that the same amounts of the compounds shown in Table 1 were used as the chlorine-containing polymerizable compound and the epoxy compound, and octadecyl trimethyl ammonium chloride (3 g in Example 6, and 2 g in Example 7) was added to polyoxyethylene lauryl ether (25g).

The results are shown in Table 1.

#### Example 8

The same procedure as in Example 1 was repeated except that the same amount (150g) of  $\text{C}_4\text{F}_9\text{CH}_2\text{CH}_2\text{OCOCH}=\text{CH}_2$  was used instead of FA, and sodium carbonate

(0.9 g) instead of sodium hydrogen carbonate (0.7 g) was used to adjust the PH to 8.

The results are shown in Table 1.

#### Example 9

The same procedure as in Example 1 was repeated except that the same amount (150g) of  $C_4F_9CH_2CH_2OCOCCH=CH_2$  was used instead of FA, vinyl chloride was omitted, the amount of stearyl acrylate was increased to 100 g, and sodium carbonate (0.6 g) instead of sodium hydrogen carbonate (0.7 g) was used to adjust the PH to 8. The results are shown in Table 1.

#### Example 10

The same procedure as in Example 1 was repeated except that the same amount (150g) of  $C_2F_5CH_2CH_2OCOCCH=CH_2$  was used instead of FA, vinyl chloride was omitted, the amount of stearyl acrylate was increased to 100 g, and sodium carbonate (0.6 g) instead of sodium hydrogen carbonate (0.7 g) was used to adjust the PH to 8. The results are shown in Table 1.

#### Comparative Example 1

The same procedure as in Example 1 was repeated except that vinyl chloride, the epoxidized soybean oil and sodium hydrogen carbonate were omitted, and octadecyl trimethyl ammonium chloride (5 g) was added to polyoxyethylene lauryl ether (25 g). The results are shown in Table 1.

#### Comparative Example 2

The same procedure as in Example 1 was repeated except that the epoxidized soybean oil and sodium hydrogen carbonate were omitted. The results are shown in Table 1.

#### Comparative Example 3

The same procedure as in Example 1 was repeated except that the epoxidized soybean oil was omitted, and sodium hydroxide (0.2 g) instead of sodium hydrogen carbonate (0.7 g) was used to adjust the PH to 9. The results are shown in Table 1.

#### Comparative Example 4

The same procedure as in Example 1 was repeated except that sodium hydrogen carbonate was omitted. The results are shown in Table 1.

#### Comparative Example 5

The same procedure as in Example 1 was repeated except that the epoxidized soybean oil was omitted. The results are shown in Table 1.

#### Comparative Example 6

The same procedure as in Example 1 was repeated except that sodium hydrogen carbonate was omitted and the amount of the epoxidized soybean oil was 10.7 g. The results are shown in Table 1.

#### Comparative Example 7

The same procedure as in Example 1 was repeated except that the epoxidized soybean oil was omitted and the amount of sodium hydrogen carbonate was 10.7 g. The results are shown in Table 1.



Table 1

	Chlorine-containing polyimide compound	Epoxy compound	Weakly basic compound	Initial pH Sealing	Solvent		Initial	After one month at 50°C				Yellowing (Initial)
								H-3 Water repellency	H-3 Chloro- form repellency	Strong/Mechanical Stability	Chemical Stability	
Ex 1	Vinyl chloride	Epoxidized soybean oil	Sodium hydrogen carbonate	7	Yes	-	5	5	4	Good	Good	Good
Ex 2	Vinyl chloride	Epoxidized linseed oil	Sodium hydrogen carbonate	7	Yes	-	5	5	4	Good	Good	Good
Ex 3	Vinylidene chloride	Epoxidized soybean oil	Sodium hydrogen carbonate	6	Yes	-	5	5	4	Good	Good	Good
Ex 4	Styryl acrylonitrile	Epoxidized soybean oil	Sodium hydrogen carbonate	7	Yes	-	4	4	3	Good	Good	Good
Ex 5	Vinyl chloride	Epoxidized soybean oil	Sodium hydrogen carbonate	8	Yes	-	5	4	5	Good	Good	Good
Ex 6	Vinyl chloride	Epoxidized linseed oil	Sodium hydrogen carbonate	7	Yes	Yes	5	4	5	Good	Good	Good
Ex 7	Vinylidene chloride	Epoxidized soybean oil	Sodium hydrogen carbonate	6	Yes	Yes	5	4	5	Good	Good	Good
Ex 8	Vinyl chloride	Epoxidized soybean oil	Sodium carbonate	8	Yes	-	4	3	4	Good	Good	Fair
Ex 9	$\text{C}_6\text{F}_5\text{CH}_2\text{CH}_2\text{OOCOCOC}_6\text{F}_5$	Epoxidized soybean oil	Sodium carbonate	8	Yes	-	5	4	5	Good	Good	Fair
Ex 10	$\text{C}_6\text{F}_5\text{CH}_2\text{CH}_2\text{OOCOCOC}_6\text{F}_5$	Epoxidized soybean oil	Sodium carbonate	8	Yes	-	5	3	4	Good	Fair	Good
Com Ex 1	-	-	-	5	Yes	Yes	2	1	2	Good	Good	Good
Com Ex 2	Vinyl chloride	-	-	3	Yes	-	3	2	Cannot be tested	Poor	Poor	Poor
Com Ex 3	Vinyl chloride	-	Sodium hydroxide	9	Yes	-	5	4	2	1	Poor	Poor
Com Ex 4	Vinyl chloride	Epoxidized soybean oil	-	4	Yes	-	5	5	4	3	Good	Fair
Com Ex 5	Vinyl chloride	-	Sodium hydrogen carbonate	8	Yes	-	5	5	4	3	Good	Fair
Com Ex 6	Vinyl chloride	Epoxidized soybean oil	-	4	Yes	-	5	5	4	3	Good	Fair
Com Ex 7	Vinyl chloride	-	Sodium hydrogen carbonate	9	Yes	-	4	3	4	3	Good	Poor

Table 1 shows that the combination of the epoxy compound (such as epoxidized soybean oil and epoxidized linseed oil) and the weakly basic acid (such as sodium hydrogen carbonate and sodium carbonate) can give good mechanical stability or chemical stability and good water- and oil-repellency, while none or only one of the epoxy compound or the weakly basic acid cannot give good mechanical stability or chemical stability and good water- and oil-repellency. The aqueous water- and oil-repellent dispersion of the present claims has excellent properties.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 10/27 2008

  
Takashi Enomoto